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ESTIMATION OF ACTIVITY IN WASTE PACKAGES - A SURVEY OF METHODS

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ESTIMATION OF ACTIVITY IN WASTE PACKAGES - A SURVEY OF METHODS

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ABSTRACT

Nine nuclear facilities in Canada and the United States were surveyed by telephone to determine their current methods for assaying the radionuclide content of packages of solid low-level radioactive wastes. Also, the international literature was surveyed to determine current and proposed methods for estimating the radionuclide content of waste packages. A bibliography of relevant reports and articles has been prepared. Two assay methods are reviewed: (1) the method of assigning a gross Curie content based on an external dose measurement; and, (2) the method of estimating the Curie content of specific radionuclides based upon external dose measurements combined with waste stream characterization or gamma spectrum analysis.

RÉSUMÉ

Une enquête a été effectuée par téléphone auprès de neuf installations nucléaires au Canada et aux États-Unis afin de déterminer les méthodes qu'elles utilisent pour mesurer le contenu en radionucléides des contenants de déchets solides de faible activité. On a aussi consulté les documents internationaux afin de déterminer les méthodes actuelles et proposées pour établir le contenu en radionucléides des contenants de déchets. On a préparé une bibliographie des rapports et articles pertinents. Deux méthodes d'essai sont examinées : 1) celle qui consiste à attribuer un contenu Curie brut fondé sur une mesure externe de la dose; et celle qui consiste à établir le contenu Curie de radionucléides spécifiques en utilisant des mesures externes de la dose combinées avec la caractérisation des déchets ou l'analyse du spectre gamma.

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1. Introduction

The management of solid radioactive wastes has become more restrictive as the volume of waste produced by the nuclear industry has grown. Both safety and economics are contributing to changes in the methods and procedures for handling, shipping, storing and disposing of solid radioactive wastes.

By far the largest volume of waste arises from routine housekeeping operations around nuclear facilities where there is radioactive contamination. In general, this waste contains very little long-lived radioactivity. However, any waste from a contaminated area must be treated as potentially radioactive. This includes materials such as floor coverings, paper, plastic sheeting, rubber gloves, mops, cleaning materials, rags,etc.

A particular characteristic of this type of waste is the limited knowledge concerning its radionuclide content. The material is normally collected and packaged on the basis of gamma dose measurement which assures safety in its immediate handling, but provides little information on the radionuclide content.

Management strategies for the longer term storage and disposal of low-level radioactive waste require a knowledge of the hazard and the life-time of the hazard. For example, the waste may contain a radionuclide with a longer life than that of the storage or disposal facility. Thus there is a need to know the specific radionuclide content of solid radwaste packages, in order that management options such as segregation, short and long-term storage or disposal can be considered. Clearly this can affect both the economic and safety aspects of radioactive waste management.

This study has been undertaken as a global review of the current and proposed methods for estimating the radionuclide content of waste packages. The survey consisted of two parts: a telephone survey of selected Canadian and American nuclear facilities and an international literature survey. Sections 3 and 4 of this report deal with the site and literature surveys, respectively.

There are two general approaches to the estimation of the radionuclide content of waste packages. The most common is a gross determination of the radioactive content by conversion from an external gamma dose measurement. This can be supplemented by analysis of the radionuclide distribution in the different waste streams to obtain a better estimate of the specific radionuclide content of a package. The alternative approach is the measurement and analysis of the gamma spectrum of the package's external field and thus the identification of the package constituents. Several papers addressing these methods were selected from the bibliography for review in Section 5.

The following section gives a general review of radioactive waste management and the generation of low-level radioactive wastes.

2. Radioactive Waste Management

The radioactive waste produced in nuclear power plants and other facilities must be managed so that the hazard is contained and isolated from people and the environment. This study is concerned with the assay of solid low-level wastes and the assay methods used. Since these are dependent upon the types of treatment, packaging and disposal strategies used, the following is an outline of waste management practices which directly affect the requirements and accuracy of the assay methods.

2.1 Types of Radioactive Wastes

In Canada the nuclear industry produces radioactive waste from six principal areas of activity:

(1) The mining and milling of uranium and thorium ore.

(2) The fabrication and reprocessing of nuclear fuel including initial enrichment.

(3) The operation and maintenance of commercial nuclear power plants.

(4) The operation of research reactors and other facilities at laboratories and universities.

(5) Institutional and industrial use of radioisotopes.

(6) Decontamination and decommissioning of nuclear facilities and laboratories.

In some countries, such as the U.S., wastes are also produced in the production of nuclear weapons and related development activities.

The primary focus of this study is the solid waste produced at nuclear power plants, which is packaged for storage or disposal, as well as at the research laboratories of Atomic Energy of Canada Ltd. At these facilities radionuclides are produced by both nuclear fission and neutron activation.

For management purposes radioactive wastes are categorized on the basis of handling and disposal criteria. These criteria include the Becquerel content, life-times and type of radioactivity resulting in the following generally accepted categorizations (e.g. Reference 12):

(1) High-level wastes (HLW) - Waste from the spent fuel stream of the nuclear fuel cycle which contains transuranic elements and fission products. The material contains long-lived radionuclides, is highly radioactive, and must be cooled.

(2) Low-level wastes (LLW) - The bulk of waste produced in the 'housekeeping' of nuclear facilities which contains a small amount of long-lived radioisotopes and a larger volume of short-lived radioisotopes and lightly contaminated materials. It requires no shielding for handling and storage.

(3) Where there is significant radioactivity in the low-level waste, it is often characterized as intermediate level waste (ILW) and it requires special handling. An example would be ion exchange resins used for chemical treatment, and decontamination.

(4) Transuranic wastes (TRU) - Waste containing a significant amount of alpha-activity which cannot be managed as LLW.

Nearly all of the radioactivity produced in a nuclear power plant is contained within the fuel elements and contributes to the high-level waste stream. Since a very small fraction of the fuel sheaths fail in normal operation leading to contamination of the primary coolant system and its auxiliary systems, there is a contribution to the low and intermediate level waste streams, through cleanup, decontamination and leakages.

Impurities, corrosion products and other particulates in the heat transport and moderator systems become radioactive from neutron activation. These radionuclides also contribute to the low and intermediate level waste streams. Ion exchange resins and filters used to clean the reactor process systems can have substantial beta and gamma activity and therefore represent an intermediate level waste requiring special handling.

Inevitably, the rooms and structures around certain reactor systems become contaminated. Moreover, all waste material from normal activity in these areas must be treated as contaminated. This leads to a large volume of low-level radioactive waste much of which is only mildly radioactive or uncontaminated.

Routine reactor wastes include charcoal filters, housekeeping materials, ion-exchange resins and filters, as well as irradiated components from service and maintenance. Ontario Hydro has reported (Ref. 1) the relative amounts of these types of wastes at CANDU power plants. They find the volumes of low and intermediate level solid wastes for each reactor to be typically;

(1) low level	-	1000 m^3/a
(2) resins and filters	-	50 m^3/a
(3) irradiated components	-	100 m^3/a

In addition to the above, decontamination wastes are generated whenever major repairs are made.

2.2 Current Management Practices for Low-Level Waste Handling

Ontario Hydro and Atomic Energy of Canada Ltd. have described their handling of low and intemediate level radioactive waste in several papers (Refs. 2 - 9). These are typical of other Canadian facilities.

Typical procedures for handling low-level wastes at U.S. nuclear power plants are described in Reference 10. Reference 11 gives a more comprehensive description of overall waste inventories and characteristics in the United States.

Other countries with advanced nuclear industries use a similar general approach to the handling of low-level wastes (Ref. 12).

2.2.1 Collection of Low-Level Waste

Procedures for collecting low-level waste at a CANDU power plant are outlined in References 1 and 4. All waste must be monitored for safe handling by personnel.

General housekeeping waste is typically collected in plastic waste bags and sorted according to the treatment procedures used at the facility. This includes segregation for compaction and incineration.

Because the general nature and origin of routine low-level waste streams are known, and because it is not practical or necessary for purposes of interim storage to identify the radionuclide content of these packages, external assay techniques have been developed for their characterization.

Intermediate level wastes, which are mainly resins and filters, must be handled with shielding and therefore are treated as a separate well-defined waste stream. The radionuclide content of these wastes is normally available by sampling and analysis.

Information on handling procedures at U.S. nuclear plants is given in Reference 10.

2.2.2 Packaging of Low-Level Waste

Low-level waste is generated in a wide variety of physical forms. As a consequence, standard packaging can only be used to a limited extent. The common standard package is the 55 gal. or 210 l drum. However, in special cases, boxes must be built to accommodate items such as irradiated components. Liquid wastes must be immobilized before they are added to the solid waste stream.

The method used to assay the waste packages must therefore accommodate a variety of different sizes and shapes of containers.

The efficient handling of low-level solid waste is made more difficult because of its large volume. Therefore, volume reduction by compaction (baling) and incineration is commonly practiced. These treatments increase the waste density and specific activity which then affects the assay for radionuclide content. Reference 7 reports waste characterization studies after volume reduction by incineration.

International practices for treating low and intermediate level wastes are summarized in Table 1. This shows the similarity of methods used to package and handle wastes in different countries.

2.2.3 Disposal of Low-Level Waste

The long-term waste management strategy being followed influences the packaging requirements for low-level solid wastes. If the waste is to be placed in a permanent disposal site, the packaging must be such that it will survive the life-time of the hazard. Thus, with this disposal method, the radionuclide content of the packages needs to be known. Where an interim

managed storage option is selected, it is not essential to characterize the radionuclide contents of the packages until they are placed in a permanent disposal site. Current international disposal practices vary and are summarized in Table 2.

When a permanent disposal method is selected, the alpha-activity of low-level waste must be considered because of the very long half-lives and specific risks involved. Alpha bearing wastes are segregated for disposal separately from the low-level disposal site. Tables 3 and 4 show current international practice for treating and disposing of alpha wastes. It should be noted that nuclear power plants do not produce large volumes of low-level wastes containing alpha emitters.

TABLE 1. INTERNATIONAL PRACTICES USED FOR TREATING LOW-LEVEL AND INTERMEDIATE-LEVEL RADIOACTIVE WASTES (Ref. 12)

	Volume					Immobilization		
	Evaporation	Ion Exchange	Precipitation	Compaction	Incineration	Cement	Bitumen	Polymer
Argentina				*	*		*	
Austria			*		*	*		
Belgium	*	*	*	*	*	*	*	*
Brazil								
Bulgaria	*	*	*				*	
Canada	*	*		*	*		*	*
China	*		*			*	*	*
Czechoslovakia	*	*	*	*			*	
Egypt					*	*		
Finland	*	*		*		*	*	
France	*	*	*	*	*	*	*	*
German Dem. Rep.	*	*	*	*			*	
Germany, Fed. Rep. of	*	*		*	*	*		
Hungary	*	*	*			*		
India	*	*	*	*	*	*	*	*
Italy	*	*	*	*	*	*	*	
Japan	*			*	*	*	*	
Korea, Rep. of	*	*	*	*		*	*	
Netherlands	*	*	*	*		*		
Pakistan								
Poland	*	*	*	*			*	
Romania	*	*	*	*	*	*		
South Africa						*		
Spain				*	*	*		
Sweden						*	*	
Switzerland					*	*	*	
UK	*	*	*	*	*	*		*
USA	*	*	*	*	*	*	*	*
USSR	*	*	*	*			*	
Yugoslavia	*	*	*			*		

TABLE 2. INTERNATIONAL PRACTICES USED FOR MANAGEMENT OR LOW-LEVEL AND INERMEDIATE-LEVEL RADIOACTIVE WASTES (Ref. 12)

Country	Disposal Practice				
	Surface Storage	Shallow Burial	Geological Repository	Well Injection	Ocean Dumping
Argentina	*				
Belgium	P				P,PP
Bulgaria	*				
Canada	*	*			
China		*			
Czechoslovakia	*				
Egypt		*	*		
Finland			P		
France	*	*			
German Dem. Rep.			*		
Germany, Fed. Rep. of			*		
Hungary		*			
India	*	*			
Italy	*	*			
Japan	*	P			P
Korea, Rep. of		*			
Netherlands					P,PP
Poland	*				
Romania	*				
South Africa		*			
Spain			*		
Sweden			*		
Switzerland			P		P,PP
UK		*			P,PP
USA		*		*	PP
USSR	*	*			
Yugoslavia	*				

P: Proposed.
PP: Past practice.

TABLE 3. INTERNATIONAL PRACTICES USED TO TREAT AND IMMOBILIZE ALPHA WASTES (Ref. 12)

Country	Treatment			Immobilization		
	Decontamination	Incineration	Size Reduction	Cement	Bitumen	Other
Belgium		*		*	*	*
France	P	*	P	*	*	*
Germany, Fed. Rep. of	*	*	*	*		
India		P	P			
Italy		P			*	
Japan		P	P			
UK	*		P	*		
USA	P	*				P
USSR		*	*	*	*	

*: Practice in use.
P: Practice is proposed.

TABLE 4. INTERNATIONAL FACILITIES USED IN DISPOSAL OF ALPHA WASTES (Ref. 12)

Country	Interim Storage	Geological Disposal	Deep-Well Disposal
Belgium	*	P	
China	*		
France	*	P	
Germany, Fed. Rep. of	*	P	
India	*	P	
Italy	*	P	
Japan	*	P	
UK	*	P	
USA	*	P	
USSR	*	P	*

*: Practice in use.
P: Practice is proposed or facility under construction.

2.3 Transportation of Low-Level Radioactive Wastes

Radioactive waste may be moved from the generating site to a storage or permanent disposal site. Its shipment is governed by the regulations respecting the transportation of dangerous goods (Ref. 43). Atomic Energy of Canada Ltd. will accept low-level waste for disposal on a commercial basis (Ref. 13). They require packaging and labelling in accordance with IAEA standards (Ref. 14) and the AECB's Transport Packaging of Radioactive Materials Regulations (Ref. 44). The IAEA standards have wide international acceptance.

The shipment of wastes creates another need for information with respect to the radionuclide content of waste packages. The contents of the packages must be known in the event of an accident. Thus, when transportation is involved, the assay method must provide a listing of the specific radionuclides contained in the package and their chemical form.

3. Survey of Nuclear Facilities

The initial phase of this project was a telephone survey of the operators of radioactive waste management of facilities in Canada. Included in the survey were the three Canadian utilities with nuclear generating stations and Atomic Energy of Canada Ltd. which operates major research laboratories. They were queried on their current practice and methods in assaying the radioactivity contained in low-level waste packages. The results are reported in Section 3.1 below.

Following the Canadian survey, four U.S. utilities were surveyed as to their current practice in estimating the radionuclide content of waste packages. The results of the survey are reported in Section 3.2 below. It should be noted that supplemental information on current U.S. practice can be obtained from the recent study by the Electric Power Research Institute (Ref. 10).

Although all radionuclides originate from fission and activation, there are differences in the waste streams between Canadian plants and their U.S. counterparts. Two of these are:

(1) The use of heavy water in the reactor process system of CANDU plants promotes operation with minimum leakage but produces more tritium.

(2) Because of the at-power refuelling capability of CANDU plants, they are normally operated with a lower burden of radioactivity in the heat transport system.

3.1 Canadian Facilities

In Canada low-level radioactive waste is being stored until permanent disposal options are evaluated and developed (Ref. 8). The waste is being treated in most cases by either compaction/baling or incineration to reduce storage volumes.

The primary concern with the radioactivity contained in these packages is for personnel safety, hence, the gamma dose rate is measured for safe handling of the waste packages. These measurements are made with standard gamma ray survey meters.

The method for handling the waste in the context of overall management practice is selected on the basis of measured dose rate and/or waste stream.

A complete characterization of the radionuclide content is not a routine procedure. However, the gross activity is estimated routinely to provide for an inventory of radioactivity being placed in waste storage.

The gross activity of the waste is based on a dose rate to Becquerel content conversion factor. These conversions are normally derived from the method of Bowman and Swindle. (Refs. 24,25) Atomic Energy of Canada Ltd. have reported (Ref. 7) using an inferential method based on an experimental relationship between the measured gamma ray field and the activity of five year old fission products.

External dose rate measurements only detect the presence of gamma emitting radionuclides. The presence of beta and alpha emitting nuclides cannot be detected with simple external measurements. Atomic Energy of Canada have reported (Ref. 8) that they divert waste, which is suspected of being contaminated with alpha-activity, from the low-level waste treatment centre to the high level waste stream.

Work has been done at the Chalk River Laboratories (Ref. 7) to develop correlations between the relative amounts of alpha, beta and gamma activities. However, there were no reports of correlations of this type being used routinely.

3.2 U.S. Facilities

In the U.S. any licensee who transfers radioactive waste to a land disposal site or to a licensed waste collector is required by 10 CFR 20 (Ref. 26) to comply with 10 CFR 61 (Ref. 27). In particular they must specify the amount of each radionuclide in the waste as summarized in Appendix A.

All of the facilities surveyed are specifying the radiological content of their waste packages. Their approaches are based on an external gamma dose measurement and a correlation with a nominal distribution of radionuclides in each waste stream.

The Electric Power Research Institute (Ref. 28) generalized the Bowman and Swindle method for generating dose to Becquerel (Curie) content conversion factors. This allowed for the different decay energies of the possible radionuclides. Therefore, provided the waste generator knows the content of his waste stream, the measured gamma dose rate can be converted to a Curie content of specific radionuclides.

The facilities surveyed are using the method developed by the Electric Power Research Institute or their own variant. The general approach is the same regardless of reactor type.

It is common in U.S. practice to use different dose rate measurements for the characterization of the waste and for radiation protection. The reason for this is largely economic which is as important a factor as safety in the development of assay methods. Overly conservative estimates of the radioactivity contained in packages places unnecessary costs on transportation and disposal. Moreover, this leads to premature filling of the disposal site which is undesirable because of the difficulties in licensing new sites.

To determine the dose rate for conversion, some utilities subtract the background dose rate. Furthermore, they do not always select the maximum measured dose rate since they want a measurement which is representative of the sample as a whole. This requires some judgement in the monitoring process. For example, in a package of low level waste, the only significant activity may be from one item in the mixture of materials. It would be a gross conservatism to assign that level of activity to the entire mass of waste. Thus, an average of dose measurements may be more appropriate in many cases.

4. Literature Survey

An international literature survey was initiated in parallel with the survey of Canadian and American utilities (Section 3). The on-line computerized data bases of the Canadian Institute for Scientific and Technical Information at the National Research Council were used. These data bases can be accessed through the CAN/OLE System on which Atlantic Nuclear Services maintains an account.

The following data bases were searched:

(1) CAS77/82/86 - Chemical Abstracts

CA SEARCH covers all aspect of chemistry and chemical engineering. Journals, monographs, patents, conference proceedings, theses, technical reports and government publications are monitored for references to new findings about applications, chemical substances, materials, procedures, reactions, techniques and theories.

(2) EI - Computerized Engineering Index

The EI database covers all aspects of engineering including chemical, civil, electronic, industrial and mechanical engineering and mining. References to geology, material on new products, news items, patents, physics, pure chemistry or theoretical methematics are excluded. Over 3,500 primary sources are covered: journals, reports and special publications of societies, research institutions, industry, government agencies and universities, theses, standards and monographs. Individual conference papers are included from 1970- July 1982.

(3) EIM - Engineering Meetings

EI Engineering Meetings provides in-depth coverage of 2,000 technical conferences held each year worldwide, and their contribution of 100,000 papers. The database includes references to each individual paper as well as the conference itself.

(4) EPD - Electric Power Database

The EPD database contains summaries of R&D projects of interest to the electric power industry in the U.S. and Canada. Included are over 1,200 projects provided by the Canadian Electrical Association. Subjects include hydroelectric power, fossil fuels, nuclear power, customer utilization, R&D support, transmission, distribution, economics, personnel, advanced power systems, stations, area development and environmental assessment.

(5) INIS - International Nuclear Information System

INIS is produced by the International Atomic Energy Agency which is composed of 50 countries and international agencies. The database covers aspects of peaceful application of atomic energy. INIS monitors journals, theses, conference proceedings, monographs, reports and patents.

Most of the databases include the technical report literature and conference papers. In the nuclear industry these are often more important sources of current information than are published papers in scientific journals. This is especially true in areas where the primary activity is conducted by plant staff rather than academic researchers.

Searching the five databases resulted in a preliminary bibliography of fifty-eight citations. From this preliminary list, the search was expanded in two ways:

(1) Individuals noted to have published one or more relevant papers were contacted in order to obtain their latest information and publications.

(2) References in the preliminary list of articles were followed up.

In total approximately 250 papers and reports were identified as meriting consideration for the survey. A review of all these articles is far beyond the limited budget and schedule for the project. However, a bibliography of the citations has been compiled and is included as Section 8 of this report.

A small number of these documents were reviewed within the scope of the project and are listed as references in Section 7.

4.1 International Aspects of the Survey

The databases selected for computer searching included the international literature and returned international citations. Examples of information obtained from the search and direct correspondence are given in References 15-23.

As can be seen from the bibliography in Section 8, most of the literature on assay techniques is from the U.S. However, the literature on overall waste management policies and proposals is more uniformly distributed internationally.

In the United States, a waste generator must comply with 10 CFR 20 and 10 CFR 61 if the waste is to be transported to a storage or disposal site. The current low-level waste disposal sites are land burial and are intended for a hazard containment mission time of up to 500 years. Therefore, for both economic and safety reasons the radiological contents of the waste must be characterized.

Not all countries plan to use shallow-burial for low-level wastes. For example, Sweden and Switzerland (Refs. 20,23) plan to use mined sites which will provide a longer containment life.

The greater emphasis on assay methods for low-level waste packages in the United States is perhaps a reflection of the different stage of evolution of waste disposal policies.

4.2 Equipment for Monitoring and Waste Management

A variety of instruments and services are available for the characterization of low-level radioactive wastes. These range from supply of simple survey meters, through volume reduction, and analytical services, to complete waste management services responding to USNRC regulations.

In general, with the exception of instrument supply, most of these services reside in the United States. This is primarily because Canadian Nuclear operators tend to do the work "in house".

A typical selection of instrumentation and services available is as follows:

It should be noted that this list is by no means exhaustive.

4.2.1 Instrumentation and Equipment

Canada

Rayonics Scientific Inc.
Downsview, Ontario

Canadian distributors for Victoreen Inc. portable survey meters as well as health physics related equipment.

United States

Canberra Industries Inc.
Meridian CT

Automated gamma scanning system for waste drum asay.

Jordan Nuclear Company
Los Angeles Ca.

Various radiation detectors survey meters and dosimeters.

Stock Equipment Company
Chagrin Falls, Ohio

Dry waste compaction equipment for LLRW.

International

Lemer & Cie
France

Low background rotating shielded cell for drummed waste monitoring measurement by gamma spectroscopy programmed for continuous (sequential) operation. (Designed to meet requirements of 10 CFR 61.)

4.2.2 Special Services

Canada

None identified.

United States

Babcock and Wilcox
Lynchburg, VA.

Volume reduction (compaction) services as well as short-term storage of LLRW offered through a centalized (Parks Township, PA.) facility.

Associated Technologies Inc.
Charlotte, N.C.

Custom design, fabrication testing and startup of volume reduction and solidification systems for LLRW.

Technical Seminars Inc.
Great Neck, NY

Training programs in LLRW management.

4.2.3 Waste Management Services

The following companies offer a "full" range of services including most or all of the following:

Analysis, system design, software, volume reduction and solidification, transportation, storage, incineration and acquisition of all regulatory approvals and documentation. All are located in the U.S. with the exception of BWB Engineering of West Germany.

ALARA Engineering Inc.
Salisbury, MA.

Atcor Engineering Systems Inc.
Avon, CT.

Pacific Nuclear Systems Inc.
Federal Way, Washington State

Science Applications International Corporation
Rockville, Maryland

Note: This company seems to provide the most comprehensive and complete service in LLRW management of those surveyed, up to and including the assumption of responsibility on behalf of the client.

Waste Management Group Inc.
Crompond, NY.

Note: This company offers similar services to SAIC including a computer Code 'RADCOR' for the characterization, classification and documentation of packaged rad waste.

International

BWB Engineering GmbH
West Germany

Commercial literature related to the above suppliers has been received and reviewed but is not included in this report.

5. Review of Assay Methods

For the purpose of this review the methods for assaying the radioactivity in waste packages have been divided into two general categories:

(1) Methods in Practice

and

(2) Methods being Developed.

The methods in practice are those identified in the survey of Canadian and U.S. facilities (Section 3). The methods being developed are those identified from the literature survey (Section 4).

5.1 Methods in Practice

All of the methods in practice are based on a conversion of the measured dose rate to activity content of the package expressed in Becquerels or Curies. These methods can be further subdivided into two groups:

(1) methods for determining the gross activity of a package without specification of the radionuclides present.

(2) methods which combine a reference radionuclide characterization of the waste streams with the gamma dose rate measurements including scaling factors.

5.1.1 Dose Conversion Factors

5.1.1.1 Method of Bowman and Swindle

Bowman and Swindle (Refs. 24 and 25) developed a look-up procedure for converting measured dose rates external to the package into Curie content. Their conversion formula applies only for right-cylinders and is expressed as:

$$Q = KR$$

Where R is the dose rate in mR/h measured at the surface of the package in its mid-plane (see Figure 1), K is a shielding function dependent upon geometry, density and macroscopic cross-section of the waste material packaged and Q is the activity in mCi.

The calculation of the conversion factor is based on five simplifying assumptions:

(1) zero wall thickness of the container

(2) unit buildup factor - In the determination of source strength from dose measurements, the buildup factor accounts for forward scattering of gamma rays. The buildup factor is the ratio of the total dose rate at the detector to the dose rate at the same point if the gamma rays were only absorbed in the shielding material.

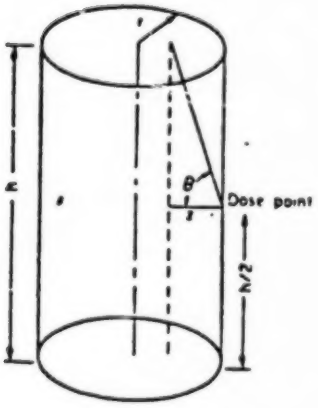

Figure 1: Cylindrical Model Representing the Radioactive Waste Container by Bowman and Swindle (Ref. 24, 25)

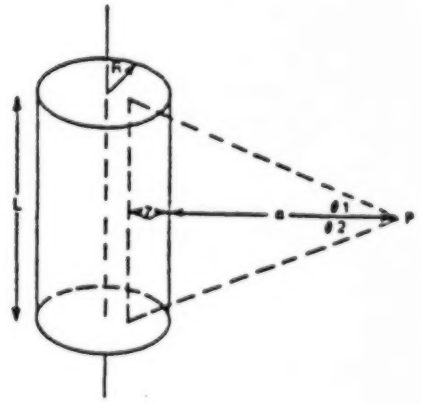

Figure 2: Cylindrical Model Representing the 55 gal. Drum Radioactive Waste Container by Lan, Kinard and Hough (Ref. 29)

(3) the dose measurement is made at the surface of the container

(4) the conversion is based on 0.5 MeV gamma rays

(5) the macroscopic cross-section was selected between carbon and water.

With these assumptions Bowman and Swindle present figures for K as a function of waste density and package diameter. They also give a table for correcting the conversion factor for gamma ray energies other than 0.5 MeV.

Applying the conversion formula requires the following input data:

(1) equivalent cylinder dimensions

(2) weight of package

(3) mean gamma ray energy.

After calculating a radius for an equivalent right cylinder and the density, K can be determined from the tables. Also, if knowledge of the representative gamma energy is available, a table is included to correct the conversion factor.

5.1.1.2 Method of Lan, Kinard and Hough

Lan, Kinard and Hough (Ref. 29) noted the industry belief that conversion factors calculated by methods such as that of Bowman and Swindle were conservative by up to a factor of 100 and developed a refined model.

The model improved upon the Bowman and Swindle model in the following respects:

(1) Both a right cylinder and a rectangular box were considered as package shapes.

(2) The dose rate is measured at a point 3 feet from the mid-point of the container and geometrical attenuation is included (see Figure 2). This improves the accuracy of the dose rate measurements over the contact measurement of Bowman and Swindle.

(3) The shielding of the container is included using the actual thickness and the appropriate material absorption coefficient. Bowman and Swindle neglect the container wall.

(4) Buildup factors are included for shielding by the container material. Bowman and Swindle assume unity.

(5) The effective gamma ray energy is to be determined from smears taken in numerous plant areas and included in the shielding and self-absorption calculations. The Bowman and Swindle calculation uses a mean gamma ray energy of 0.5 MeV.

(6) Lan, Kinard and Hough include the number of photons per decay which is not done by Bowman and Swindle.

Application of the method proceeds by the following sequence of steps:

(1) Assemble Input Data:

(i) Calculate effective gamma energy based on analysis of smears.

(ii) Determine container shield material and thickness.

(iii) Determine the shield absorption coefficient for the effective gamma energy.

(iv) measure the dose rate at a distance 3 feet from the midpoint of the container.

(2) Calculate the Source Self-Absorption

(3) Calculate the Dose Rate

(4) Calculate the Curie Content

Although this method is a refinement of the Bowman and Swindle method, it requires detailed calculations to be done for every package. Clearly it can be used in practice only if parametric tables are prepared.

5.1.1.3 Method of EPRI

The Electric Power Research Institute undertook to develop a simplified and accurate method for use by plant personnel in estimating the Curie content of radwaste packages from external radiation field measurements (Ref. 28).

A computer program (DOSCON) was developed to run on a desk top computer with 64K RAM. It allows for the following range of routine plant waste shipments:

(1) Twenty-two radionuclides

(2) Twenty-two different waste package types and geometries

(3) A wide range of filling materials

(4) Dose rates measured at 1 or 3 foot distances from the container.

The input to the code includes the identification of the container type, filling material, relative radionuclide distribution of the waste and the mid-plane dose rate measurement.

The dose rates are measured in the mid-plane of the package to simplify detector geometry (see Figures 1 and 2). A contact measurement of the dose will give a result which is largely determined by the concentration of radioactivity. A measurement at a distance (say 3 feet) gives a dose that is

largely determined by the volume of radioactive material. The use of dose rates determined at a distance has to be tempered with the need to obtain a high enough gamma flux for an accurate measurement.

The survey meter used to measure the dose rate at a point detects the gamma rays of all energies at the point and indicates a dose rate for absorbed energy. Some of the gamma flux detected will not have the energies of the gamma rays emitted from the radionuclides in the waste package. In passing through the material of the waste package, the gamma rays will be absorbed and scattered. The scattered gamma rays will be of lower energy but still contribute to the dose. The buildup in dose at the detector is described by the factor

$$B = \frac{\text{total dose rate at the detector}}{\text{unscattered gamma ray dose rate at the detector}}$$

An interesting feature of the code is the capability to account for decay and it calculates the individual nuclide concentrations at the time of shipment. Radionuclides included in the code are listed in Table 5. The radionuclide content of a low-level waste sample at a Canadian facility is given in Table 6 for comparison.

As a part of the verification of the computer code it was compared with the Bowman and Swindle calculation to determine the effect of buildup in the gamma flux. The DOSCON code predicted buildup factors of about 2 where Bowman and Swindle had assumed 1. Including the correct buildup factor reduced the estimated Curie content by 20-40%.

Allowing for dose rate measurements at a distance from the package gives better resolution of the source and should give a more accurate result.

The DOSCON code permits a wide range of package types and contents to be assayed in a relatively routine manner and with reasonable accuracy. However, it only considers the gamma activity since it is a dose conversion factor calculation.

The program is based on the following relations for the dose rate D and the total number of Curies, C:

$$D = \sum_{i=1}^{n} (d_i c_i)$$

$$C = \frac{D}{\sum_{i=1}^{n} (d_i f_i)}$$

where

$$c_i = f_i C$$

and f_i is the fraction of the nuclide, i, and d_i is its dose rate per Curie.

The DOSCON code was verified against the measured millicurie content of a cemented 55-gallon drum of concentrated wastes. The measured total activity was 25.2 millicuries and the DOSCON calculations based on 1 and 3 foot dose measurements were 22.9 and 24.0 millicuries.

The verification of the DOSCON code shows that the mathematics of the conversion are fairly accurate. Nevertheless, the overall accuracy of the estimate of the Becquerel content of a package remains dependent on the two measurements:

(1) the external dose rate measurement for the individual package which is input for the DOSCON code,

and,

(2) the characterization of the waste stream, from which the package was taken, according to the relative concentration of different radionuclides which is input for the DOSCON code.

Table 5. NUCLEAR DATA INCLUDED IN THE DOSCON CODE (Ref. 30)

A	Element	Half-Life
51	Cr	27.704d
54	Mn	312.7d
57	Co	270.9d
58	Co	70.80d
59	Fe	44.63d
60	Co	5.27y
65	Zn	244.4d
95	Zr	64.02d
95	Nb	35.06d
103	Ru	39.35d
103M	Rh	56.12m
106	Ru	368.2d
106	Rh	29.92s
113	Sn	115.1d
113m	In	1.658h
124	Sb	60.20d
125	Sb	2.77y
131	I	8.040d
131M	Xe	11.84d
134	Cs	2.062y
136	Cs	13.16d
137	Cs	30.17y
137M	Ba	2.552m
140	Ba	12.789d
140	La	40.22h
141	Ce	32.5d
144	Ce	284.3d
144	Pr	17.28m

Table 6. GAMMA-RAY EMITTING RADIONUCLIDES IDENTIFIED IN CRNL LLW (Ref. 7)

Radionuclide	Halflife (days)	Percentage of Total Activity (%)
Nb-95	35	30.9
Zr-95	64	16.4
Co-60	1924	15.0
Cr-51	28	10.7
Cs-137	10,987	7.1
Ce-144	284	5.0
Zn-65	244	3.5
Ce-141	33	2.6
La-140	1.7	1.9
Ag-110m	253	1.7
I-131	8.1	1.1
Ru-103	40	0.9
Co-58	71	0.7
Ba-140	13	0.6
Cs-134	752	0.5
Ru-106	369	0.5
Fe-59	45	0.5
Sb-124	2.7	0.1
Mn-54	313	0.1

5.1.1.4 Method of CRNL

A non-invasive technique for assaying low level radioactive waste has been developed at the Chalk River Nuclear Laboratories (CRNL) and placed in-service at their Waste Treatment Center (Ref. 9, 45). Details of the design and theory of the monitor have not been published. The following is a summary of the monitoring concept as outlined in Reference 9.

(1) A waste shipment is characterized by both a gross gamma survey of all packages and a gamma spectrum analysis of selected packages.

(2) The wastes are characterized prior to volume reduction and standardized packaging. Gamma ray absorption and the effects of density variations are stated to be minimal.

(3) Each package is monitored for gross gamma activity. If the gamma flux exceeds a pre-set level, the gamma spectrum of the package is then analyzed.

(4) The total activity in the shipment is obtained from the gross gamma measurements with the gamma spectrum analysis being used to prorate the total among the specific radionuclides.

(5) The quantities of alpha and beta-emitting radionuclides in the waste shipment are estimated from correlations with gamma-emitting nuclides in the particular type of waste.

(6) The background level of activity in the monitoring area is measured and subtracted from the package measurements.

(7) Both sets of gamma-ray monitors are calibrated with reference sources imbedded in material typical of the waste shipment.

There was no information on the verfication of the system available for review.

5.1.2 Sampling of Waste Streams

As discussed in Section 5.1.1.3, the overall accuracy of the dose conversion method is limited by the accuracy of the radionuclide distribution. The distribution of gamma active nuclides can be measured relatively easily from waste samples using a Ge Li detector and analyzing the gamma spectrum.

It is not practicable to analyze every sample or indeed to analyze samples from waste streams on a frequent basis. Thus, the accuracy of the distribution function becomes a function of the sampling procedures and quality control of the waste management program.

Factors to be considered include the following:

(1) selection of representative sample points and sample frequency for segregated waste streams.

(2) identification of the origin of the waste packages being assayed

(3) recognition of the need to sample the waste stream when there is reason to expect a change in the nuclide distribution.

It is much more difficult to determine the distribution of beta and alpha emitting radionuclides (Ref. 30). Often facilities must have the analysis done off-site (Ref. 31 and 32) and the turn-around time can be fairly long. When the analysis is done off-site, the Quality Assurance program must include the off-site laboratory.

There exist computer codes (Ref. 33) which classify and document the individual packages of radwaste so that an integrated history docket can be maintained. This approach greatly assists the management and control of the waste generated and its proper characterization.

5.1.3 Scaling Factors

The external dose rate measurements for waste packages characterize only the gamma emitting radionuclides. There is no external package measurement of the alpha and beta emissions. Thus, alpha and beta active radionuclides contained in the package must be determined from a correlation with the gamma active radionuclides as derived from waste stream sample analysis.

The requirements of 10 CFR Part 61 for characterizing non-gamma radioactivity are given in APPENDIX A.

The method used for characterizing the beta and alpha activity involves the assignment of scaling factors based on association with gamma emitters such as Co-60 and Cs-137. However, this method suffers from the difficulty in

measuring the scaling factors of the radionuclides in the waste stream samples. To generate a larger data base, the sample data from different facilities have been combined. It has been observed (Ref. 34) that these generic scaling factors can be as much as four orders of magnitude different from the actual scaling factors at a particular plant.

Plant specific scaling factors must be developed (including waste stream specific factors) to obtain the nominal factor of 10 as required by 10 CFR 61.

5.1.4 Stored Waste

The preceding discussion of methods in use to characterize the radionuclide content of packages only applies to waste being currently generated. Waste generated in the past and placed in storage without characterization is lacking in information with respect to radionuclide distribution. This will become a problem when the waste is to be taken from storage and placed in a permanent disposal site. Since the manual sampling of the waste packages is not practicable, nondestructive techniques will need to be developed for identifying specific nuclides (Ref. 9 and 35). The bibliography contains reports on work in this area.

5.2 Methods Being Developed

There are a number of methods under development to improve the characterization of wastes where dose conversion methods as discussed in Section 5.1.1.3 cannot be applied. The principle areas of development effort include:

(1) non-destructive assay of gamma emitting radionuclides (Ref. 9, 36).

(2) characterization of beta emitting radionuclides by external measurements (Ref. 36).

(3) characterization of alpha contaminated or TRU wastes (Ref. 36-42).

The assay of gamma emitting radionuclides can be done by using detectors to analyze the energy spectrum of the external gamma field. Systems that have been developed for this purpose are described in References 9 and 36. They offer the real possibilty of non-destructive characterization of wastes. However, the waste density and homogeneity of the activity become complicated by incineration and compaction. It may be possible to obtain density information from differential gamma absorption. A detailed review of these systems is beyond the scope of this project.

Brodzinski (Ref. 36) has proposed a system which incorporates the detection of Sr-90 by an external bremstrahlung measurement. This is the only technique found for detecting beta activity by non-destructive monitoring. Again, analysis of the physics of the method and its accuracy and utility are beyond the scope of this project.

There is abundant literature on the subject of assay for the alpha radioactivity contained in waste. The principle methods involve using a

neutron source to induce fission with detection of gamma rays or delayed neutrons. A sample of the research and development effort is contained in References 36-42.

6. Conclusions

A total of nine Canadian and U.S. facilities were surveyed for their current practices in the characterization of low-level waste packages. Also, an international literature survey was conducted and a bibliography of citations was prepared.

Methods being practiced by the waste generators for assaying low-level waste have been reviewed. The complete characterization of low-level waste packages requires an integrated waste management program. The actual detection methods, such as gamma dose rate measurements, require supplementary information and control procedures to fully document the specific gamma emitting radionuclides contained in the waste.

Non-destructive assay techniques for gamma, beta and alpha activity were not reviewed because of the limited scope of the project. There is a substantial literature on the development of these methods.

The literature was found to contain very little information on the comparison of methods. There was one specific case of a comparison of the Doscon code (Ref. 30) with the assumption made by Bowman and Swindle (Ref. 25) regarding the buildup factor for the gamma dose. There is a very definite need for a numerical comparison of different methods.

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APPENDIX A

REQUIREMENTS OF 10 CFR PART 61

A.1 Regulation 10 CFR Part 61

In the U.S. the land disposal of low-level radioactive waste is regulated by 10 CFR Part 61 (Ref. 1). The objectives of the proposed regulations are (Ref. 2):

(1) To ensure that the movement of radioactivity does not result in a calculated dose rate exceeding 25 mR/year to an individual at the site boundary, or cause the EPA drinking water standard to be exceeded at the nearest public drinking water supply.

(2) To assure that an inadvertent intruder does not receive an exposure exceeding 500 mR/year.

To meet the disposal objectives four classes of waste are defined:

(1) Class A Segregated Wastes - Wastes to be segregated from other wastes because they do not have to be packaged with the stability of longer lived wastes. They contain activity of such low level or short half-life that contamination of water supplies is not a concern.

(2) Class B Stable Wastes - Wastes that will decay to non-hazardous levels in the 100 year period that access to the burial site will be controlled.

(3) Class C Intruder Wastes - Wastes which will not harm the public or an intruder over a 500 year period. The Quantity, life time and packaging must be appropriate for this target.

(4) Unacceptable for Low-Level Burial - Wastes which contain sufficient long lived nuclides to exceed the Class C Intruder Waste limit.

The nuclides for which 10 CFR 61 specifies limits and their nuclear properties are summarized in Tables A-1 and A-2.

A.2 References

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Table A-1
10 CFR 61 WASTE CLASSIFICATION ACTIVITY LIMITS (Ref. 2)

	uCi/cm^3		
	Class A	Class B[a]	Class C
Long Lived			
C-14	0.8	NA	8
C-14 in activated metal	8.0	NA	80
Ni-59 in activated metal	22.0	NA	220
Nb-94 in activated metal	0.02	NA	0.2
Tc-99	0.3	NA	3
I-129	0.008	NA	0.08
Alpha emitting transuranics with half-lives >5 years	10[b]	NA	100[b]
Pu-241	350[b]	NA	3,500[b]
Cm-242	2,000[b]	NA	20,000[b]
Short Lived			
Total of nuclides with half-lives <5 years	700	(c)	(c)
H-3	40	(c)	(c)
Co-60	700	(c)	(c)
Ni-63	3.5	70	700
Ni-63 in activated metal	35	700	7,000
Sr-90	0.04	150	7,000
Cs-137	1	44	4,600

a) There is no Class B category for wastes exceeding the Class A limit for long lived nuclides. Such wastes are automaticlaly Class C or are unacceptable for shallow burial. For wastes containing a mixture of nuclides, the sum of the fractions of limits must be less than one. The appropriate limits must all be taken from the same column in the table.

b) Units are nanocuries per gram.

c) There are no specified limits for these nuclides in these clases. Practical considerations such as heating effect, external package radiation on shipping, or maximum specific activity determine the maximum concentration for these nuclides.

Table A-2

NUCLEAR CHARACTERISTICS (Ref. 2)

Nuclide	Max. Beta Energy MeV (% abundance)	Gamma Energy MeV (% abundance)
12.35 year H-3	0.0186 (100)	No gamma
5730 year C-14	0.155 (100)	No gamma
7.5×10^4 year Ni-59	Electron capture	No gamma[a]
100.1 year Ni-63	0.0659 (100)	No gamma
5.27 year Co-60	0.318 (100)	1.172 (100)
2.03×10^4 year Nb-94	0.473 (99)	0.7026 (98) 0.8711 (100)
28.82 year Sr-90	0.546 (100)[b] 2.288 (100)[b]	No gamma
2.14×10^5 year Tc-99	0.292 (100)	No gamma[c]
1.57×10^7 year I-129	0.150 (100)	0.040 (7.5)
30.17 year Cs-137	0.512 (94) 1.176 (6)	0.6616 (89.9)[d]

a) Detectable only by Co-59 x-rays and small amount of internal bremstrahlung

b) From 64.1 hour Y-90 daughter activity

c) Gamma transitions exist but less than 1×10^{-5}% abundance

d) From 2.55 minute Ba-137m activity